REPORT DOCUMEI ATION PAGE

AFRL-SR-AR-TR-08-0240

1. AGENCY USE ONLY (Leave blank)	2. REPORT DATE March 31, 2008	3. REPORT TYPE AND DATES COVERED Final Report 01/01/2005 - 12/31/2007

4. TITLE AND SUBTITLE
Fundamental role of grain boundaries: meso-scale simulation and measurement

5. FUNDING NUMBERS
AFOSR Collaborative Grant Nos.
FA9550-05-1-0068 and
FA9550-05-1-0088

6. AUTHOR(S)
Robert H. Wagoner
Brent L. Adams

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)
The Ohio State University, Dept. of Materials Science & Engineering
2041 College Rd., Columbus, OH 43210

Brigham Young University, Dept. of Mechanical Engineering
Provo, UT 84602

8. PERFORMING ORGANIZATION REPORT NUMBER
GRT8351200/ 60000956

9. SPONSORING / MONITORING AGENCY NAME(S) AND ADDRESS(ES)
Dr. Joan Fuller
AFOSR Program Manager, Metallic Materials
875 North Randolph St.
Suite 325, Room 3112
Arlington, VA 22203-1768

303 10. SPONSORING / MONITORING AGENCY REPORT NUMBER
Air Force Office of Scientific Res
(50010105)

11. SUPPLEMENTARY NOTES
N/A

12a. DISTRIBUTION / AVAILABILITY STATEMENT
Approved for public release; distribution is limited.

12b. DISTRIBUTION CODE
N/A

13. ABSTRACT (Maximum 200 Words)
Collaborative Projects FA9550-05-0068 and FA9550-05-0088 were aimed at solving one of the outstanding problems in the mechanical behavior of metallic materials: understanding, quantifying, and predicting the role of grain size and grain boundary character on strength and ductility. This is an essential component required for the design of high-temperature alloys to resist stresses. Current models for polycrystals address only the grain texture (i.e. the statistical orientation of crystal lattices), not the presence or character of grain boundaries, nor the size of the grains.

Novel simulation methods and materials characterization techniques were developed and verified. Noteworthy progress and results may be summarized as follows:

- Identification of the underlying physics of grain-size effects in metals and alloys.
- First quantitative prediction of Hall-Petch slopes from first principles (without introducing arbitrary length scales).
- First comparison of measured and independently predicted lattice curvatures at grain boundaries. (Good agreement was discovered: within 20%.)
- First simulations and presentations of dislocation density distributions within grains
- Introduction of a new single-crystal constitutive representation that is more accurate and uses fewer parameters than existing equations. (Outstanding agreement was found with measurements.)

14. SUBJECT TERMS High-temperature structural materials, Orientation Imaging Microscopy, Multi-Scale Modeling, EBSD, Hall-Petch, Grain size, Grain boundary, Lattice curvature, Single crystals, Polycrystals, Geometrically necessary dislocations, Statistical dislocations

15. NUMBER OF PAGES
13

16. PRICE CODE N/A

17. SECURITY CLASSIFICATION OF REPORT Unclassified	18. SECURITY CLASSIFICATION OF THIS PAGE Unclassified	19. SECURITY CLASSIFICATION OF ABSTRACT Unclassified	20. LIMITATION OF ABSTRACT Unclassified and Unlimited

FUNDAMENTAL ROLE OF GRAIN BOUNDARIES:

MESO-SCALE SIMULATION AND MEASUREMENT

Submitted by co-PI's

Robert H. Wagoner, George R. Smith Chair
The Ohio State University
Dept. of Materials Science and Engineering
2041 College Road
Columbus, OH 43210-1178
Phone: 614-292-2079
Fax: 614-292-6530
E-mail: Wagoner.2@osu.edu

Brent L. Adams, Dusenberry Professor
Brigham Young University
Dept. of Mechanical Engineering
Provo, UT 84602
Phone: 801-422-7124
Fax: 801-422-0516
E-mail: b_l_adams@byu.edu

TABLE OF CONTENTS

FUNDAMENTAL ROLE OF GRAIN BOUNDARIES: MESO-SCALE SIMULATION AND MEASUREMENT

ABSTRACT

Collaborative Projects FA9550-05-0068 and FA9550-05-0088 were aimed at solving one of the outstanding problems in the mechanical behavior of metallic materials: understanding, quantifying, and predicting the role of grain size and grain boundary character on strength and ductility. This is an essential component required for the design of high-temperature alloys to resist stresses. Current models for polycrystals address only the grain texture (i.e. the statistical orientation of crystal lattices), not the presence or character of grain boundaries, nor the size of the grains.

Novel simulation methods and materials characterization techniques were developed and verified. Noteworthy progress and results may be summarized as follows:

- Identification of the underlying physics of grain-size effects in metals and alloys.
- First quantitative prediction of Hall-Petch slopes from first principles (without introducing arbitrary length scales).
- First comparison of measured and independently predicted lattice curvatures at grain boundaries. (Good agreement was discovered: within 20%.)
- First simulations and presentations of dislocation density distributions within grains
- Introduction of a new single-crystal constitutive representation that is more accurate and uses fewer parameters than existing equations. (Outstanding agreement was found with measurements.)

TECHNICAL BACKGROUND

Multi-scale models are gaining momentum in many fields as computers become faster, and finer structures become accessible experimentally. An effective multi-scale model of dislocation-based metal plasticity has potential impact for many important applications such as enabling polycrystalline material design by prediction of the interaction of dislocations with microstructural features. This is an important aspect of high-temperature material performance.

Recent advances in metal plasticity have been focused in two widely separated length scales:

1. Texture analysis (macro scale) revolutionized metal plasticity in the last 15 years by predicting the form of plastic anisotropy of textured metals. This greatly reduced the requirement to perform onerous numbers of experiments to determine this property for each alloy. Texture analysis, however, does not register or respond to local structures, i.e. dislocations, particles, grain boundaries. Rather, it averages or homogenizes continuum properties over many crystallite orientations by sacrificing local equilibrium, compatibility, and input from local structure.

2. Molecular dynamics (nano-scale) and related experimental techniques (e.g. TEM, nanoindentation) reveal discrete single-dislocation properties in reference volumes of a few thousands of atoms. This work is important for understanding the behavior of nano-scale structures, where discrete dislocation properties govern the properties. These advances have had limited impact on continuum constitutive equations for typical structural metals and alloys where larger scales and large numbers of interacting dislocations are involved.

The principal obstacle facing a predictive model connecting these length scales has been the large numbers of defects involved (typically 10^{12}-10^{16} dislocations/m^2), or, conversely, the wide disparity of scales to be linked (10^{-10} m for a typical burgers vector versus 10^{-2} m for a part or component, or, 10^{-5} m for a typical grain in a polycrystal). This disparity of scales renders a direct/ traditional multi-scale model of metal plasticity computationally intractable. The methods developed in these projects allow treating statistical numbers of dislocations at the meso scale while preserving the essential characteristics of discrete dislocations and avoiding heavy computational overhead. Detailed verification was an equally daunting task that has been accomplished, as outlined below.

The pervasive effect of grain boundaries on material properties is readily seen in the well-known Hall-Petch relationship [1,2] between grain size D and yield strength σ_y:

$$\sigma_y = \sigma_o + kD^{-\frac{1}{2}} \tag{1}$$

were σ_o is the stress to move dislocations in the absence of grain boundaries, k is the Hall-Petch slope. This relationship provides a good, but by no means perfect [3-5], description of material behavior for a wide range of metals and alloys, and over a wide range of grain sizes, but there is no satisfactory quantitative model to predict Equation 1, particularly for bcc materials which have much higher slopes than fcc ones.

PROJECT OBJECTIVES

The joint 2005 AFOSR project (FA9550-05-0068 and FA9550-05-0088) was aimed at developing two novel techniques and applying them cooperatively to create a unique approach to the problem of predicting quantified interactions between dislocations and grain boundaries, thus providing a verified method to understand and predict the Hall-Petch effect in metallic polycrystals.

Two novel and challenging methods were sought (see Figure 1):

1) Simulation: A meso-scale finite element method (FEM) based on inhomogeneous dislocation generation, annihilation, and accumulation was to be formulated, implemented and tested. It was then to be used to predict the dislocation content inside grains in a polycrystal, along with resulting mechanical properties.

2) Measurement: Orientation Imaging Microscopy (OIM), invented by PI Adams [6], was to be extended to resolve lattice curvature within grains and near grain boundaries. Extremely fine polishing techniques had to be developed and validated, as well as techniques for handling the large amount of information necessary.
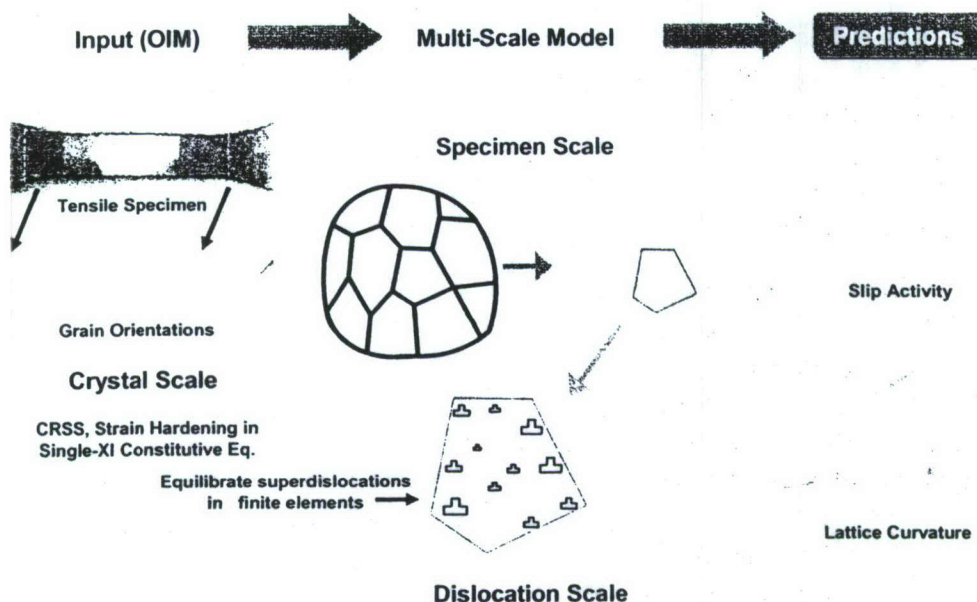
Figure 1: Schematic of Multi-Scale Model and its verification.

In order to understand the role of grain boundary character and grain size, comparisons of simulated and measured quantities were proposed: lattice curvature (related to inhomogeneous elastic strain and dislocation content) and macroscopic stress-strain curves. A small number of physical parameters needed to be established by these comparisons; two scalars of particular importance being identified: τ^*, the maximum grain boundary stress resisting dislocation transmission, and χ, the fraction of dislocation generation rate that represents mobile dislocation density.

RESULTS

The key developments in simulation and OIM have been accomplished, and the methods demonstrated and verified using Fe-3%Si as a model material.

Simulation Results

After derivation of the complex equations needed for the meso-scale simulation, simple numerical tests were conducted to test the robustness of the method. For example, a classical pile-up problem was solved using the simulation and the solution was shown to be independent of the mesh configuration, except for the obvious refinement in spatial resolution available for finer FE meshes, Figure 2.

The first predictions of dislocation densities and lattice curvatures for polycrystals were based on standard single-crystal constitutive equations [7,8] as implemented in texture analysis. These results were clearly nonphysical. It became clear that the major problem was using the fictitious single-crystal constitutive equations derived for texture calculations (SCCE-T's). SCCE-T's conceal the physical effects of grain boundaries by fitting

macroscopic measurements, which depend on grain boundaries, to SCCE-T's via texture analysis, which ignores grain boundaries. In effect, all of the grain boundary physics is contained in the spurious single-crystal representation.
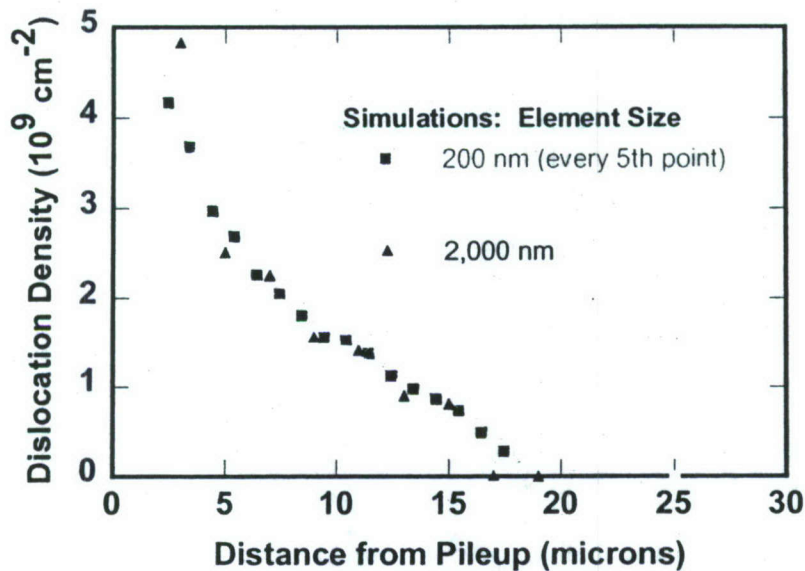

Figure 2 – Pile-ups simulated with the proposed multi-scale model: 1-D pile-up results reproduce an analytic solution [14] independent of element size.

To address this shortcoming of the texture approach to single-crystal constitutive behavior, dislocation-based single-crystal constitutive equations (SCCE-D's) were derived starting from formulations in the literature. These forms are not only more realistic, they require fewer undetermined parameters, 4 (vs. 6 for SCCE-T's).

To test the accuracy of SCCE-T's and SCCE-D's, the deformation of single-crystals of fcc and bcc alloys was simulated using FEM. In each case the equations were fit to single slip data in the literature, and in each case the predictions for multiple slip were compared. Figure 3 shows that the SCCE-D's are an order of magnitude more accurate, which illustrates the critical importance of grain boundaries to the macroscopic properties of polycrystalline metals and alloys.

The parametric test shown in Figure 4 was also used to assess the variation of macroscopic flow stress with grain size, Figure 5, by enlarging and reducing the size of the problem. (Texture analysis predicts no dependence, as does meso-scale simulation by setting $\tau^*=0$ or $\tau^*=\infty$.) The form of the experimental Hall-Petch relationship is predicted directly from single-crystal properties. *This is the apparently first such prediction of the Hall-Petch relationship, for any physically realistic model involving dislocations.* Note, however, that the Hall-Petch slope is quite small, as might be expected for a two-grain / columnar model. A significant part of future work will focus on extending the simulation techniques for larger assemblies of grains.
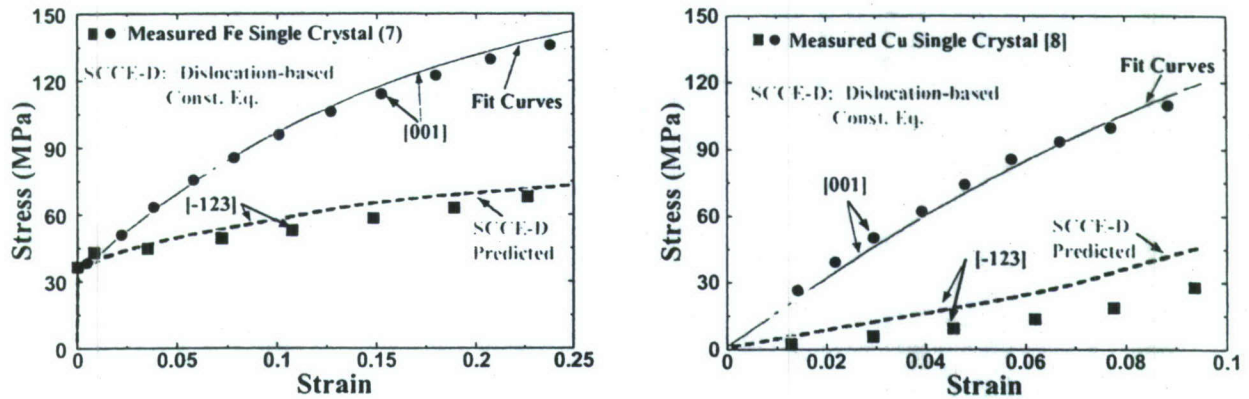
Figure 3 – Comparison of experimental results for single crystals with finite element simulations based on texture-based constitutive equations (SCCE-T) and dislocation-based constitutive equations (SCCE-D). The figure on left is for Fe (bcc); the one on the right is for Cu (fcc). Both kinds of equations were fit to tensile results aligned to 001. [9-12]
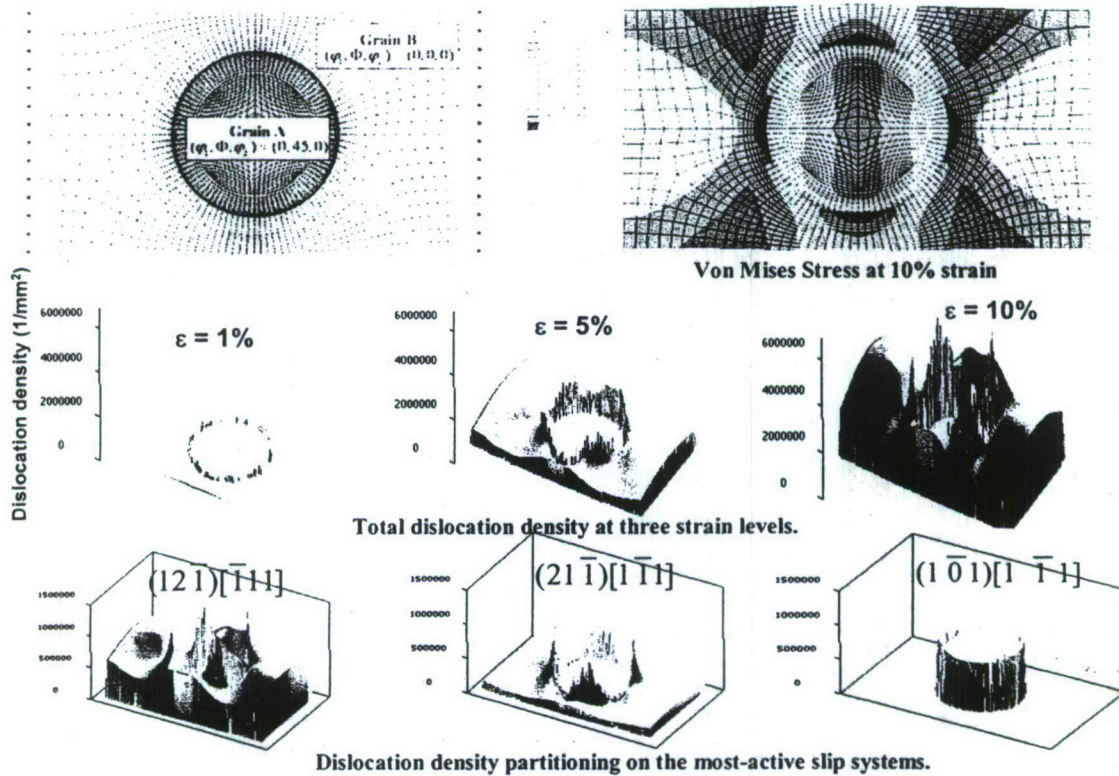

Von Mises Stress at 10% strain

Total dislocation density at three strain levels.

Dislocation density partitioning on the most-active slip systems.

Figure 4 – Two-scale simulation of a grain within a grain, boundary obstacle strength $\tau^*=5\sigma_{CRSS}$, lattices misoriented by 45°. The stress contours (as affected by elastic incompatibility and dislocation content) appear at the upper right. The simulated development of dislocation density throughout the problem as a function of strain appears in the middle row of figures. The lower row of figures shows the partitioning of dislocation density on slip systems at 10% strain.

The first true comparison of simulations and measurements of lattice curvature for a real polycrystal is shown in Figure 6. The meso-scale FEM simulation predicted, based on true single crystal behavior, the inhomogeneous slip activity, compatibility, and mechanical equilibrium in a dislocation-statistical sense without introducing an arbitrary length scale. A tensile specimen with 9 columnar grains was OIM analyzed for grain orientations, then deformed in tension, and then reanalyzed for lattice curvature. In parallel, the configuration was simulated and the lattice curvatures predicted. The results show remarkable agreement, with the maximum lattice curvatures within about 20%. However, some grain boundaries were predicted to have high curvature whereas no such curvature was observed. This is presumably related to the simplifying assumption used that the boundaries are infinite obstacles to dislocation transmission or absorption.

Note: Figures 4 and 6 represent the apparently first-ever predictions of spatial dislocation densities (or lattice curvatures) for an applied deformation in a polycrystal taking into account elastic and plastic incompatibility at grain boundaries.
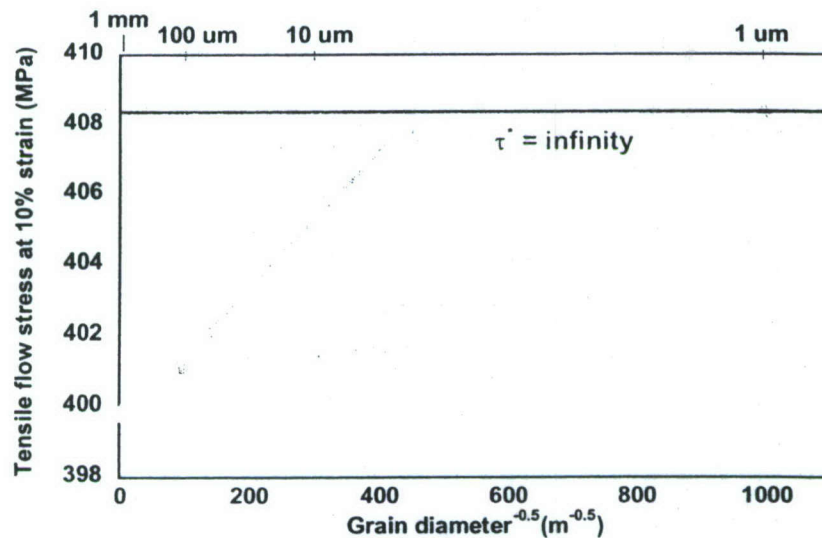

Figure 5 – Hall-Petch prediction of model shown in Figure 4. The size of the specimen and the embedded grain are varied proportionally.

Figure 6 – First example of simulated (left) and measured (right) lattice curvature for a coarse-grained, columnar tensile Fe-3%Si specimen after straining to 20% strain. The simulated and measured maximum lattice curvatures agree within 20%.

Characterization Results

OIM techniques, particularly the critical polishing procedure, were shown capable of resolving lattice curvature at a limit well below that needed, depending on the OIM step size (Figure 7). As demonstrated by Figures 6, fine scale OIM to resolve lattice curvature is a powerful tool for characterizing the defect fields that develop in polycrystalline materials subjected to plastic deformation. The results from the first two project years demonstrated that ~98% of all lattice orientation measurements are resolved to within $\theta^* = 0.5^o$ of true orientation. Thus, fine scale OIM at a step size d ($d \geq 20\ nm$), is capable of resolving the spatial fields of lattice curvature, κ, to a resolution limit of $\kappa \approx \theta^*/d \approx 5 \times 10^5\ rad/m$.

The resolution already attained corresponds to dislocation densities of $\sim 10^{16}\ m^{-2}$, without elastic strain gradients. Cold-worked materials typically have *average* dislocation densities in excess of 10^{12} - $10^{16}\ m^{-2}$, with near-boundary regions and triple point densities that may exceed $10^{17}\ m^{-2}$. (Peak densities of $\sim 10^{18}\ m^{-2}$ have been observed near some grain

$$\kappa_{ij} = -\alpha_{ij} + \frac{1}{2}\delta_{ij}\alpha_{kk} - e_{ilk}\varepsilon^e_{jk,l}$$
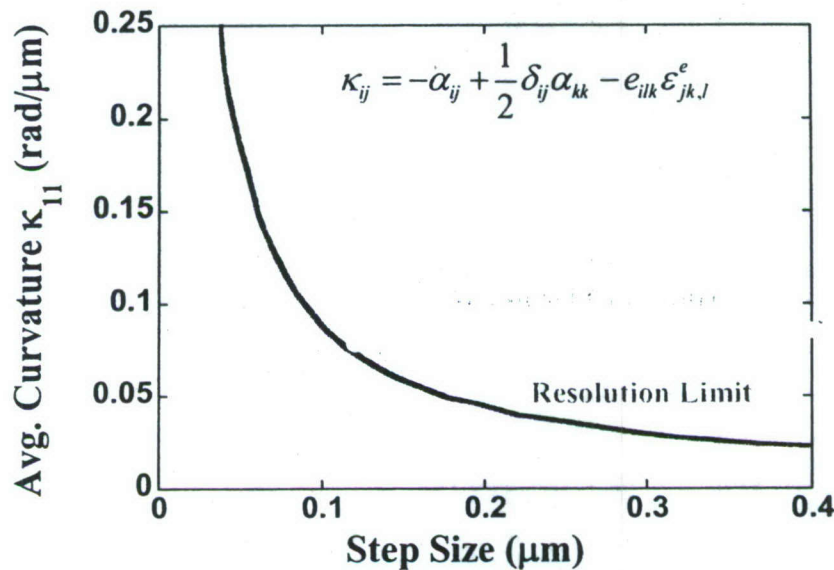
Figure 7 – Resolution limit of OIM vs. measured curvature in Fe-3%Si specimens.

boundaries of the low alloy steel of this project.) Clearly, since curvatures and dislocation densities that lie above the average levels are resolvable, improvements in the resolution limit for lattice curvature, if achievable, would be highly desirable. Improved curvature resolution is the experimental focus of continued work.

FUTURE DIRECTIONS

Collaborative Projects FA9550-05-0068 and FA9550-05-0088 are finished, except for completing papers in progress. The results are promising and novel; they represent the only ones of their kind to have appeared. However, there are limitations and uncertainties that should be addressed in order to enable practical and reliable application of these techniques to improve the design of materials. A future proposal to AFOSR will focus on these issues.

Future work will need to address the principal limitations of the current approach, as follows:

- The simulation represented in Figure 6 (9 grains) required 7.2 hours of CPU time on a standard 2.8 GHz PC. The algorithm must be improved in terms of efficiency and more powerful computers must be utilized to extend such calculations to 50-100 grains. (Hundreds of grains are usually considered sufficient in texture calculations to capture the essential physics, but the limit is not known for the new technique.) It is for this reason that many of the demonstration simulations make use of $\tau^*=\infty$; such simulations are approximately 10 times faster, but do not capture the size effect of the grain.

- While preliminary comparisons show amazing agreement, the experimental data has thus far been insufficient to quantify some key simulation parameters. Simulations and measurements for numerous specimens with varied grain size and boundary character (20-100 specimens) must be compared to obtain verified values of τ^* and χ. (Recall that τ^* is the scalar grain boundary strength that scales the local obstacle strength for each boundary depending on boundary orientation and crystal misorientation. χ is the ratio between mobile and total dislocation densities.) The simulations to date have used a reasonable estimate of $\tau^*=5\sigma_{CRSS}$ from the fcc metal literature [13] and an assumed equi-partitioning of mobile and immobile dislocations.

- There has been no direct relationship between the FEM mesh used in the simulation part of this work and the location at which OIM diffraction patterns are taken. This can lead to errors in comparisons of lattice curvatures because such values depend explicitly on the extent of the region surveyed and the distance between adjacent locations used in the calculation.

BROADER IMPACT

In addition to the applications of direct interest to the Air Force, other applications and benefits can be envisioned as follows:

Broader Benefits – Fundamentally new capabilities in computational metal plasticity and its characterization will enable material design and improve applications, e.g. metal forming. The methods developed bridge length-scale gaps between single-dislocation behavior, computational methods, and continuum plasticity for finite-strain applications without introducing arbitrary unknowns and fitting parameters. The methods may also have impact beyond metal plasticity, i.e. using the meso-scale simulation techniques derived and verified.

Enhanced Infrastructure – Orientation Imaging Microscopy, invented (and named) by PI Adams in the 1990's, is today widely used to investigate the structure and properties of a range of materials. The proposed advances in polishing and OIM techniques and software will allow direct and complete measurement of lattice curvature, closely related to GND content. For metal applications, this ability provides a quantitative method to characterize the local dislocation density content that is otherwise unavailable.

PROJECT PUBLICATIONS

Publications in Print:

1. E.R. Homer, B.L. Adams, and R.H. Wagoner, "Recovering Grain Boundary Inclination Parameters through Oblique Double Sectioning," *Metallurgical and Materials Transactions A* **38A**, 1575-86, 2007.

2. B.L. Adams, C. Landon, J. Kacher, and R.H. Wagoner, "Experimental methodology for the study of metals at large plastic strains by electron backscatter diffraction," Proceedings of the Second International Symposium Physics and Mechanics of Large Plastic Strains, St. Petersburg, Russia, 4-9 June 2007, published in *Problems of Materials Science* **52**, 51-6, 2007.

3. C.D. Landon, B.L. Adams and J. Kacher, "High Resolution Methods for Characterizing Mesoscale Dislocation Structures," *ASME Journal of Engineering Materials and Technology,* accepted for publication, 2007.

4. M. G. Lee, R. H. Wagoner, B. L. Adams: *Two-Scale Simulation of Dislocation – Grain Boundary Interactions,* Halifax, Canada, Proceedings of the 12th International Symposium on Plasticity (Plasticity 2006), pp. 283-285.

5. B.L. Adams, C. Landon, J. Kacher, *Characterization of the Mesoscale Dislocation Density Tensor Field via High-Resolution Electron Backscatter Diffraction,* Proceedings of PLASTICITY 08: The Fourteenth International Symposium on Plasticity and its Current Applications, p. 172-174, Fulton, Maryland, 2008.

6. C. Landon, B.L. Adams, D. Fullwood, J. Kacher, *High Resolution Orientation Imaging Microscopy: Obstacles and Innovations*, Proceedings of the 15th International Conference on Textures of Materials (ICOTOM 15), 2-6 June 2008, in press.

7. J. Kacher, B.L. Adams, D. Fullwood, C. Landon, *Separating Coincident Electron Backscatter Diffraction Patterns Near Interfaces*, Proceedings of the 15th International Conference on Textures of Materials (ICOTOM 15), 2-6 June 2008, in press.

Publications in Preparation:

8. M.-G. Lee, R. H. Wagoner, B. L. Adams: *Two-Scale Simulation of Dislocation – Grain Boundary Interactions: Theory and Numerical Framework*, Int. J. Plasticity (in preparation).

9. M.-G. Lee, R. H. Wagoner, B. L. Adams: *Two-Scale Simulation of Dislocation – Grain Boundary Interactions: Identification of Physical Parameters*, Int. J. Plasticity (in preparation).

10. R. H. Wagoner, M. G. Lee, B. L. Adams: *Multi-Scale Model and Verification of Dislocation – Grain Boundary Interactions*, Plasticity of Conventional and Emerging Materials: Theory and Applications, eds. A. S. Khan, B, Farrokh, NEAT Inc., 2007, pp. 100-102.

11. C. Landon, J. Kacher, B.L. Adams, D. Fullwood: *High Resolution EBSD by Cross Correlation with Simulated Patterns,* Ultramicroscopy 2008 (in preparation).

REFERENCES

1. Hall, E.O: The Deformation and Ageing of Mild Steel, <u>Proc. Phys. Soc. B</u>, 1951, vol. 64, pp. 747-753.

2. Petch, N. J.: The Cleavage Strength of Polycrystals, 1953. <u>J Iron Steel Inst.</u> 174, 25.

3. Azrt, E. 1998. Size effects in materials due to microstructural and dimensional constrains, A comparative review. Acta Met 46(16), 5611-5626.

4. Hirth, J.P. 1972. The influence of grain boundaries on mechanical properties. Met Trans 3, 3047.

5. Smith, T.R., Armstrong, R.W., Hazzledine, P.M., Masumura, R.A. and Pande, C.S. 1995. Mats Res Soc Symp Proc. 362, 31.

6. Adams, B.L., Wright, S.I. and Kunze, K., 1993. <u>Metall. Trans. A.</u> 24A,819.

7. Asaro, R.J., Rice, J.R., 1977. <u>J. Mech. Phys. Solids</u> 25, 309.

8. Peirce, D., Asaro, J.R., Needleman, A., 1982. <u>Acta Metall.</u> 30, 1087.

9. M. G. Lee, R. H. Wagoner, B. L. Adams: *Two-Scale Simulation of Dislocation – Grain Boundary Interactions,* Halifax, Canada, <u>Proceedings of the 12th International Symposium on Plasticity (Plasticity 2006</u>), pp. 283-285.

10. R. H. Wagoner, M. G. Lee, B. L. Adams: *Multi-Scale Model and Verification of Dislocation / Grain Boundary Interactions*, <u>Proceedings of the 13th International Symposium on Plasticity (Plasticity 2007)</u> (accepted for publication)

11. M.-G. Lee, R. H. Wagoner, B. L. Adams: *Two-Scale Simulation of Dislocation – Grain Boundary Interactions: Theory and Numerical Framework*, <u>Int. J. Plasticity</u> (in preparation)

12. M.-G. Lee, R. H. Wagoner, B. L. Adams: Two-Scale Simulation of Dislocation – Grain Boundary Interactions: Identification of Physical Parameters, <u>Int. J. Plasticity</u> (in preparation).

13. Z. Shen, R. H. Wagoner, and W. A. T. Clark: Dislocation and Grain Boundary Interactions in Metals, Acta Metall., 1988, Vol. 36, pp. 3231-3242.

14. Hirth, J.P. and Lothe, J. 1969. *Theory of Dislocations*. McGraw-Hill, New York. 704.